Electric dipole and quadrupole interaction assisted nonlinear optical properties of CdSe Quantum dot

SHORT SYNOPSIS

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by

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ABSTRACT

Quantum dots (QDs) have received significant attention due to their unique optical properties and potential applications in photonics, optoelectronics, and quantum computing. Our synopsis subject is dedicated to the exploration of some of the nonlinear optical characteristics of CdSe quantum dots, highlighting their distinctive attributes and the mechanisms behind nonlinear optical phenomena. Nonlinear optical effects in quantum dots differ fundamentally from those in bulk materials owing to quantum confinement effects, resulting in discrete energy levels. These effects give rise to a broad spectrum of nonlinear optical phenomena, including nonlinear absorption, harmonic generations and optical bistability. The size-dependent electronic structure of CdSe quantum dots facilitates efficient frequency conversion processes such as second-harmonic generation, optical rectification, sum-frequency generation, difference frequency generation and third harmonic generation providing opportunities for compact, efficient light sources across different spectral regions. Furthermore, the strong nonlinear optical response of quantum dots can be enhanced and customized through various strategies, such as adjusting QD size, shape engineering, surface engineering, as well as by controlling the QD composition and morphology. These approaches offer control over the nonlinear optical properties, facilitating their integration into advanced photonic and optoelectronic devices. In conclusion, the nonlinear optical properties of quantum dots offer promising applications in ultrafast optics, telecommunications, and quantum information processing. Understanding and harnessing these properties are crucial for the development of next-generation photonic technologies with improved performance and functionality.

Keywords: Intersublevel transitions, Semiconductor Quantum Dot, Second Order Optical Nonlinearity, Quadrupole Interaction

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1. INTRODUCTION

Nanomaterials have brought a noteworthy transformation to various fields throughout natural sciences and engineering. Investigating nanoscale materials is one of the greatest captivating research topics in the world of science today. Owing to its remarkable applicability in nearly every sector of our civilization, including the chemical, electrical, and optoelectronic industries, medical imaging, pharmaceutical, and sensing industries, studies on nano-structured materials are at an advanced level today. These materials, at a nanometer scale, exhibit extraordinary properties and introduce new fundamental physical phenomena. This occurs because of greater surface-to-volume ratio and the appearance of quantum phenomena affect the nanoscale (Negi et al., 2013). In primary 20th century there is critical contributions, such as Einstein's calculation of molecular diameter and the invention of the electron microscope, which played a crucial role in progressing the field of nanoscience (Akbarnavaz Farkoush et al., 2013a). However, physicist Richard Feynman from Caltech ignited an enthusiasm for nanoscale research with his groundbreaking presentation titled "There is Plenty of Room at the Bottom" during the yearly summit of the American Physical Society. During his talk, Feynman talked deep about the possibilities of manipulating objects on a small scale and highlighted the vast potential of this unexplored field (Safarpour et al., 2014a). A decade after Feynman's influential presentation, the introduction of a novel method called molecular beam epitaxy (MBE) for crystal growth which is marked as a noteworthy milestone (Jaouane et al., 2023a), enabling the production of semiconductor nanostructures (Cho and Arthur, 1975). Norio Taniguchi introduced "nanotechnology" to describe fabrication methods that controlled sizes at the nanometer level (Niculescu, 2012). A pivotal flash originated the scanning tunneling microscope for the development, and Gerd Binning and Heinrich Rohrer won the Nobel Prize for it (Toda et al., 2013).

The field of nanotechnology gained momentum with the publication (Drexler, 1981) of Eric Drexler's paper and his subsequent book, "Engines of Creation: The Coming Era of Nanotechnology" (Cheng and Yan, 2009). Following these key developments, noteworthy progress continued in the realm of nanoscience, including the invention of atomic force microscopy (Binnig et al., 1986), the discovery of fullerenes (Xu et al., 2010), carbon nanotubes, and more (Safarpour et al., 2014b).

Nanotechnology is rooted in nanoscience, focusing on the unprecedented material properties at the quantum level dependent on size. Nanoscience and technology are advancing in various domains, encompassing materials science, optoelectronics, biomedical science, and more

(Vasiliev et al., 2015). Remarkable advancements have been made in semiconductor technology, particularly in the generation of size-controlled semiconductor nanostructures (Rabanian et al., 2023a).

Nanoparticles are often invisible to the human eye, considering the smallest particle size that can be seen with the unaided eye is greater than a nanoparticle (around 0.1 mm). By conventional methods, we are unable to view objects with sizes lesser than the visible range of electromagnetic spectrum, even with optical microscopes (Niculescu et al., 2013). Subsequently, it is exceptionally challenging to conduct a direct analysis of the size, shape, and chemical composition of nanoparticles. The number of molecules or atoms in a material diminishes with decreasing size.

The key goal of this size-controlled engineering is to design devices that leverage quantum effects, resulting in unconventional electron transport and linear and non-linear optical properties across a vast spectral range, ranging from near to long-infrared wavelengths.

1.1 Semiconductor nanostructure and their significance

Semiconductors hold a crucial role in modern-day science and technology. The innovative work of Shockley, Bardeen, and Brattain at Bell Laboratories, leading to the invention of semiconductor transistors, set the stage for the practical application of semiconductors (Ozmen et al., 2009). Following this, Moore's law predicted the miniaturization of semiconductor devices, resulting in enhanced functionality, increased integration capacity, and reduced energy consumption (Wang and Mizaikoff, 2008). This miniaturization brought quantum effects to the forefront. As a result, semiconductor devices based on low-dimensional semiconductor nanostructures revolutionize the semiconductor industry. Throughout semiconductors, several characteristic length scales are intrinsic to their material properties. Notably, interatomic spacing and the free exciton Bohr radius play noteworthy roles in shaping the properties of semiconductor nanostructures (Makhlouf et al., 2021). These properties become critical when the size of semiconductor nanostructures approaches a few of these characteristic length scales. Experimentally, semiconductor nanostructures have been synthesized in various forms, including quantum wells (2D structures with 1D confinement) (Akbarnavaz Farkoush et al., 2013a), quantum wires (1D structures with 2D confinement) (Dasgupta et al., 2014), and quantum dots (0D structures with 3D confinement) (Abbasi et al., 2017). These categories are reliant on the number of reduced dimensions they retain, and they play a fundamental role in the properties and applications of low-dimensional materials.

In low-dimensional semiconductor nanostructures, whose size approaches these characteristic lengths, new physical properties emerge. Due to these characteristics, quantum mechanical effects can be observed, such as the Coulomb-blockade effect in zero-dimensional structures (Beenakker et al., 1991; Roest et al., 2003), which is categorized by conductance resonances, and the quantum Hall effect in two-dimensional electron gas (Duhan et al., 2017), which is connected to the quantization of conductance.

These effects are comprehensively related to discrete energy levels and charge quantization. The low-dimensional semiconductor nanostructures offer the ability to tailor various material properties, including electronic and optical characteristics, for advanced applications in future devices.

1.2 Quantum confinement in semiconductor nanostructures

Quantum confinement pertains to the confinement of electrons and holes throughout semiconductor materials with dimensions similar to the characteristic Bohr radius, specifically the Bohr radius of an exciton. An exciton consists of an electron-hole pair physically separated by the Bohr radius, symbolizing the distance between the electron and the hole in the conduction band and valence band respectively. (Goncalves et al., 2023a).

Excitons are categorized into two types based on the strength of Coulomb interaction between the electron and hole: Wannier excitons and Frenkel excitons. Consecutively, their binding energies are approximately 10 meV and 100 meV. Wannier excitons in semiconductors are particularly sensitive to the material's spatial geometry, and the energy of an exciton is lower than the transition energy at the band edge by its binding energy.

Semiconductor nanostructures exhibit two distinct potentials: the Coulomb interaction potential and the spherical well potential. An exciton's Coulomb interaction energy scales with ε^2, where ε signifies the dielectric constant. Meanwhile, the confinement energy of an electron-hole pair owing to the spherical potential well with a radius of R scales with $\mu R^2/\hbar^2$, where μ is the reduced mass of the electron-hole pair. The three different confinement regimes are recognized based on the relative magnitudes of confinement and Coulomb interaction energies viz; weak confinement regimes, intermediate confinement regimes and strong confinement regimes.

The weak confinement regime is when the Coulomb interaction energy is noteworthily balances the individual confinement energies of the electron and hole, this regime is called weak confinement. This applies to bulk materials and large nanoscale materials. In the weak

confinement regime, the system's physical size is much greater than the Bohr radius $\Delta >> a_B^*$ (i.e., $\Delta >> a_B^{e*}, a_B^{h*}$), where the Bohr radii can be expressed as: $a_B^{e,h*} = \frac{4\pi\varepsilon_0\varepsilon_r\hbar^2}{m_{e,h}^* e^2}$.

In the intermediate confinement regime, the Coulomb energy is no longer inconsequential associated to the confinement energy. This happens when the system's physical size (Δ) is smaller than one carrier's individual Bohr radius but larger than the other carrier's Bohr radius (i.e., $a_B^{e*} > \Delta > a_B^{h*}$). This is due to the electron having a smaller effective mass than the hole. In the case of strong confinement regime, the Coulomb energy is negligible compared to the confinement energy. This happens when the system's physical size (Δ) is much reduced than both the individual Bohr radii of the electron and the hole ($\Delta << a_B^{e*}, a_B^{h*}$. for both electrons and holes). Small nanomaterials fall into this category, where quantum confinement effects dominate the material's optical properties.

When one or more dimensions of a bulk semiconductor material are reduced to the order of 10^{-9} nanometers (nm), noteworthy changes in the material's properties and behavior occur, resulting from the quantum confinement of electrons and holes. These modified structures are called as low-dimensional structures. Quantum confinement also leads to changes in the density of electronic states throughout these low-dimensional structures compared to bulk materials (Jaouane et al., 2023b). The density of states (DOS), denoted as $\mathscr{D}(E)$, signifies the number of available energy states throughout a specific energy range per unit dimensions of real space. Multiplying the DOS by the energy interval gives the total concentration of available states throughout that energy range (Fenoll et al., 2023).

1.3 Motivation: why quantum dot?

Quantum dots provide us an opportunity to investigate semiconductor behaviors in the finite size regime. Due to quantum confinement the charge carriers in quantum dots (QDs) are confined in all three dimensions, they behave differently from quantum wires and wells. Quantum dots have many fundamentally interesting and potentially useful optical properties. The density of states in QDs resulting from three- dimensional quantum confinement is delta-like with well-separated energy levels, as compared with continuous bands in the bulk or sub bands of energy levels in semiconductor quantum wells and quantum wires (Sobhanan et al., 2023a). Quantum dots act accordingly, like artificial atoms. The optical properties of quantum dots are extremely fascinating because of their atomic-like structures. By altering the dot's size, shape, substance, and confinement potential, the energy levels can be observed at various

values (Vahdani and Ehsanfard, 2018). The optical characteristics of quantum dot nanostructures can be tailored by the use of electronic transitions at discrete energy levels. The size of dot can be altered to modify their band gap energy (Dasgupta et al., 2014), luminescence wavelength, transition probability, photoabsorption, and emission spectra. These quantum dot properties are highly helpful for the development of optoelectronic devices, including biological imaging (Caruge et al., 2008), photovoltaics (Nozik et al., 2010; Sun et al., 2007), and light-emitting diodes (Kamat, 2008; Michalet et al., 2005).

1.3.1 The quantum dot: quantum confinement in 3 dimensions

A quantum dot (QD) is a three-dimensional semiconductor structure conventionally enveloped by a wide-band gap semiconductor material. The size of this semiconductor nanostructure is similar to the Bohr radius, a_B. For carriers throughout this structure, it functions as a three-dimensional potential well. Consequently, charge carriers are confined in all three spatial directions, creating a structure often likened to an artificial atom. The term "QD" was first coined by Reed (Reed et al., 1988).

The earliest and most widely used technique for growing QDs involves their incorporation into a glass matrix (Ekimov et al., 1985). Additionally, QDs have been fabricated in colloidal solutions, a notable achievement by L. E. Brus in collaboration with Efros and Ekimov who had recently awarded by the noble prize in chemistry for their work on quantum dots (Rossetti et al., 1983; Zurita-Sa Nchez and Novotny, 2002) (Fenoll et al., 2023; Jaouane et al., 2023a, 2023b). The concept of quantum confinement was initially introduced by Efros and Efros (Rossetti et al., 1984), who noticed changes in the color of glasses as a result of variations in the size of mixed CdS_xSe_{1-x} crystals. This remark was then through an experiment confirmed by Ekimov (Cibert et al., 1986) and deliberated in terms of the color changes in semiconductor colloidal solutions by Rosetti et al.

Several studies have provided evidence of quantum confinement effects in QDs, boosting our understanding of these nanostructures. To demonstrate the quantum confinement effect, Reed et al. conducted various experiments involving electron transport through a three-dimensionally confined GaAs quantum well quantum dot (Ozmen et al., 2009).

Recent explored efforts have focused intensively on the optical properties of semiconductor QDs. These properties are instrumental in unraveling the fundamental understanding of quantum effects throughout semiconductor nanostructures. Various experimental and theoretical studies have contributed to an increasingly comprehensive comprehension of the

size-dependent electronic and optical properties of QDs (Jeong et al., 2001; Klimov et al., 2001; Yoffe, 1993).

For a detailed account of the progress made up to the end of the 20th century and the beginning of the 21st century, one can refer to (and their associated references) (Greilich et al., 2006; Marin et al., 1998), as the ongoing expansion of knowledge and advancements in this field continue to improve (Gupta et al., 2017; Mi et al., 2012; Qu and Peng, 2002; Zurita-Sa Nchez and Novotny, 2002) (Cabezas et al., 2023; Goncalves et al., 2023a; Hasaneen et al., 2023; Hien, 2022; Nasa and Purohit, 2023; Rabanian et al., 2023b).

In case of infinite potential barrier, the potential for the confinement along three dimensions will be expressed as:

$$V^{(1,2,3)}(x,y,z) = \begin{cases} 0 & 0 < x < L_x, 0 < y < L_y, 0 < z < L_z \\ \infty & otherwise \end{cases} \quad (1)$$

And Schrodinger equation reduced to,

$$-\frac{\hbar^2}{2m^*}\left(\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}+\frac{\partial^2}{\partial z^2}\right)\psi(x,y,z) = E\,\psi(x,y,z) \quad (2)$$

The explanation of equation (2) is given by the eigen function in the form of,

$$\psi_{n_x,n_y,n_z}(x,y,z) = \frac{\sqrt[2]{2}}{\sqrt{L_xL_yL_z}}\sin\frac{n_x\Pi x}{L_x}\sin\frac{n_y\Pi y}{L_y}\sin\frac{n_z\Pi z}{L_z} \quad (3)$$

The eigen function agreeing to a specific eigen energy is obtained as,

$$E_{n_x,n_y,n_z} = \frac{\hbar^2}{2m^*}\left(\frac{k_x^2}{L_x^2}+\frac{k_y^2}{L_y^2}+\frac{k_z^2}{L_z^2}\right) \quad (4)$$

For the QDs the density of states is a series of delta function and can be given in this form,

$$\mathscr{D}_{0D}(E) = 2\sum_{n_x,n_y,n_z}\delta(E-E_{n_x,n_y,n_z}) \quad (5)$$

1.4 Interaction of light and matter (Genesis of optical properties)

The interaction between light and matter is a powerful means to collect insights into the characteristics of substances, accomplished with the help of a wide array of spectroscopic techniques spanning a vast spectrum of wavelengths. This interaction forms the foundation for comprehending the optical properties of matter and constitutes various types of transitions, including absorption, emission, scattering, and ionization, resulting from both single-photon and multiphoton processes (Rabanian et al., 2023c).

During the processes for single photon, when a single electron interacts with a single photon in a system, this interaction is called as a single-photon process. It can be explained in terms of

a matrix element, denoted as $\langle f|V|i\rangle$, where $|i\rangle$ and $|f\rangle$ represent the initial and final states of the electron in system, and V indicated the coupling of the electron's interaction with the incident radiation.

During the processes for multiphoton, in situations where the intensity of light is sufficiently high, it becomes possible to observe processes that involve the absorption of multiple photons. These scenarios are defined as multiphoton processes. For example, an electron has transition from its ground state to an excited state by absorbing two or more photons. Moreover, a bound electron may absorb three photons and subsequently emit one photon with an energy which is equal to the summation of the energies of the three incident photons. This is called third harmonic generation, which happens when all three photons retain similar frequencies.

1.4.1 Electric dipole interactions

In the framework of the semiclassical theory, the interaction between an optical field and a material is defined using a multipolar expansion.

$$exp\left(i\,\vec{k}\,.\,\vec{r}\right) = 1 + \left(i\,\vec{k}\,.\,\vec{r}\right) + \frac{1}{2!}\left(i\,\vec{k}\,.\,\vec{r}\right)^2 + \ldots$$

This expansion is characterized by the Hamiltonian, given by:

$$\widehat{H} = \widehat{H_0} + \widehat{H'}$$

Here, $\widehat{H'}$ indicates the unperturbed Hamiltonian of the system, and in the absence of a radiation field it is relating to the motion of electron. Contrastingly, $\widehat{H_0}$ describes the interaction between the field and electrons.

$$\widehat{H'} = \hat{\mathcal{E}}\,.\,\vec{r}\,(e^{i\,\vec{k}.\,\vec{r}})$$

In this equation, ε is the unit polarization vector, and $\vec{k}$ is the photon's propagation vector.

Electric dipole interaction couples state with opposite parity. When dealing with far-field excitation, such as at lower optical frequencies, the first term in the multipole expansion, called the electric dipole term, results in a noteworthily stronger response compared to subsequent terms. This happens because the physical dimensions of the material are very lesser than the wavelength of the incident light, and the field's spatial variation is relatively weak. To put it concisely, the electric dipole interaction is responsible for light-matter interactions at lower optical frequencies.

In the dipole approximation, the electric dipole term can be simplified as approximately equal to $(e^{i\,\vec{k}.\,\vec{r}}) \approx 1$ and higher-order terms in the multipole expansion are conventionally neglected under these conditions.

1.4.2 Electric quadrupole interactions

At higher optical frequencies, beyond the electric dipole interaction, the electric quadrupole interaction assumes a noteworthy role in various optical properties. Contrastingly the electric dipole interaction, which states opposite parity, the electric quadrupole interaction couple states of the same parity. The second-rank tensor electric quadrupole operator, indicating its higher order compared to the electric dipole (Corer and Hodes, 1994; Ithurria et al., 2011; Milichko et al., 2013; Shreekanthan et al., 2003; Singh et al. 2014; Umar et al., 2012).

In cases, where the spatial variation of optical fields is considerably stronger, as is the situation in near-field excitation scenarios, the higher terms in the multipolar expansion must not be dismissed. For example, the subsequent terms, such as the electric quadrupole and magnetic dipole interactions, respond to the gradient of the electric field. As a result, the electric quadrupole interaction, whose intensity resembles to that of the electric dipole interaction, is incorporated into the optical interaction when high-field gradients are involved (Wassel et al., 2024).

In the framework of quantum dots (QDs), where confined states with the same symmetry are common, intersubband optical transitions are established through the quadrupole interaction, while electric dipole transitions are prohibited. These different selection rules for transitions governed by the electric dipole and the electric quadrupole interactions open up unexplored possibilities for applications in optoelectronics and photonics.

2. LITERATURE REVIEW

2.1 Transition from Bulk CdSe to CdSe Quantum Dots

Many scientific fields have benefited from the revolutionary discoveries and applications that have resulted from the development of semiconductor materials from their bulk form to nanoscale dimensions. The transition from bulk CdSe to CdSe quantum dots (QDs) is one of the biggest developments in this regard, with special qualities and exciting potential uses. This section seeks to clarify the transition from bulk CdSe to CdSe QDs by examining the structural, optical, and functional characteristics of these materials, as well as the variety of uses for which they are employed (Vahdani and Ehsanfard, 2018).

Bulk CdSe is a well-known semiconductor material with unique characteristics linked to its bandgap (~ 1.73 eV) and crystalline structure. However, the optical and electrical behaviours

of CdSe are changed when it becomes confined to nanoscale quantum dots due to quantum confinement effects (Duhan et al., 2017) (Ramaiah et al., 2001). Through size-dependent bandgap engineering, CdSe QDs display tunable optical properties, offering a range of colours with applications in photonics, biological imaging, and optoelectronics (Khomane and Hankare, 2010; Vahdani and Ehsanfard, 2018).

CdSe QDs are synthesised using a variety of methods, including organometallic approaches, solvothermal synthesis, and colloidal methods (Sobhanan et al., 2023b). These techniques provide exact control over the QDs stability and optical characteristics by affecting their size, shape, and surface chemistry. For CdSe QDs to be more stable, less poisonous, and easier to integrate into a variety of applications, surface passivation and functionalization are essential (Goncalves et al., 2023b).

There is a significant impact of quantum confinement effects on the optical characteristics of CdSe QDs, including their emission and absorption spectra. When the size of the QDs decreases, the quantum size effect causes a blue shift in the optical bandgap, which enables the tuning of emitted light across the visible spectrum. Additionally, CdSe QDs are attractive for usage in lighting, sensing, and displays due to their remarkable photoluminescence quantum yield (Hankare et al., 2004). Moreover, CdSe QDs has distinct optical and electrical characteristics which makes them the perfect choice for photovoltaic applications, where their capacity to capture light can boost solar cell efficiency (Nair et al., 1993). Furthermore, biological imaging uses its size-tunable emission features to monitor and image biomolecules in living systems at high resolution.

The manufacturing, stability, and toxicity of CdSe QDs continue to present challenges, despite significant advancements and possible uses. In order to reduce their environmental impact, surface engineering techniques and sustainable synthesis procedures are critical to addressing these issues and ensuring their broad use in many industries (Brahim et al., 2021).

There continue to be challenges with CdSe QD synthesis, stability, and toxicity, despite the tremendous progress and prospective uses. It is still imperative to address these issues in order to ensure their broad use in a variety of industries, highlighting the significance of environmentally friendly surface engineering techniques and sustainable synthesis techniques (Pisheh et al., 2017).

In conclusion, the switch from bulk CdSe to CdSe quantum dots has created a variety of options by enabling previously unachievable control over optical and electrical properties at the nanoscale. A thorough grasp of their synthesis, structure, and functionalization opens up a wide range of applications, including biomedical imaging and optoelectronics. In order to

completely realize the potential of CdSe quantum dots in improving a variety of technological sectors, future research attempts should concentrate on overcoming current problems and investigating innovative applications.

2.2 Review of CdSe quantum dots over the past decade

Over the last decade, CdSe quantum dots (QDs) have been the subject of widespread research and development beholding remarkable advancements and applications across various fields.

2.2.1 Innovation in synthesis and structure

The synthesis of CdSe quantum dots with exact control over size, shape, and surface properties has advanced significantly. Utilizing novel synthetic methods, such as thermal degradation and chemical approaches, CdSe nanostructures, including nanorods, nanoplatelets, and core/shell QDs, have been created (Yong et al., 2009). Chemical bath deposition (CBD), vacuum evaporation (Ramaiah et al., 2001), electrodeposition, spray pyrolysis (Pourjafari et al., 2012), thermal evaporation (Shreekanthan et al., 2003) and successive ionic layer adsorption and reaction (SILAR) are the most prevalent methods for creating CdSe thin films (Chen et al., 2013; Murali et al., 2004; Wassel et al., 2024). In order to improve optoelectronic capabilities of CdSe nanostructures (Choi et al., 1998), researchers have concentrated on attaining a uniform size distribution and customizing structural aspects.

2.2.2 Optical and Electronic Properties

Knowing and manipulating the optical and electrical characteristics of CdSe quantum dots has been essential to their advancement. Important areas of investigation have included their variable absorption/emission edges, high quantum yield, and size-dependent photoluminescence throughout the visible spectrum (Pourjafari et al., 2012). These nanocrystal's distinct optical characteristics are the result of quantum confinement effects, which make them excellent choices for a range of photonics, optoelectronic, and bioimaging applications (Chen et al., 2013).

2.2.3 Applications in Optoelectronics

Applications for CdSe QDs can be originate in a variety of optoelectronic devices, such as photodetectors, solar cells (Obrien and Mcaleese,1998), LEDs, and laser diodes (Nguyen,

2010) (Kalsoom et al., 2021). Being able to emit light at a variety of wavelengths and having efficient light-emitting qualities have made them highly sought-after materials for enhancing gadget performance (Barglik-Chory et al., 2003).

2.2.4 Biomedical and Sensing Applications

CdSe QDs have been thoroughly investigated in the biomedical field for their prospective use in targeted drug delivery, bioimaging, and diagnostics. They are used as fluorescent probes for labelling and imaging biological structures due to their fluorescence characteristics, outstanding photostability, and biocompatibility (Barglik-Chory et al., 2003) (Wagner et al., 2005) (Zhu et al., 2000). Their ion-sensing properties have also been researched for a range of sensing uses.

2.2.5 Challenges and Future Prospects

Despite their encouraging qualities, there are still issues, especially with regard to the toxicity of heavy metals like cadmium. To reduce toxicity concerns, surface modification and encapsulation have been the focus of efforts (Sobhanan et al., 2023a). Furthermore, research is still ongoing to improve the scalability and repeatability of CdSe QD production while maintaining their optoelectronic characteristics.

Future investigations into surface engineering, synthesis techniques, and customized applications in different industries are expected to encourage further progress in CdSe quantum dots (Djurisic and Leung, 2006). In command to enable broad use of CdSe quantum dots in commercial applications, these initiatives seek to fully realise the potential of these particles while handling current obstacles.

2.3 Linear and Nonlinear Optical Properties of CdSe Quantum Dots

2.3.1 Linear optical Properties of CdSe Quantum Dots

Investigations into CdSe quantum dots (QDs) have revealed an intriguing world of nanomaterials with remarkable linear optical characteristics. The purpose of this part is to explore the intriguing linear optical behaviour of CdSe quantum dots and provide insights into their basic characteristics, synthesis processes, and wide range of applications in many scientific fields.

When CdSe quantum dots interact with light in a linear manner, their reaction is directly proportional to the intensity of the incident light. This phenomenon is known as linear optical characteristics (Yang, 1999). These characteristics, which affect the electrical structure and optical behaviour of CdSe QDs, result from the quantum confinement effect, which is a consequence of their nanoscale size (Ramya et al., 2020). Because of their effective light absorption and charge carrier dynamics, CdSe QD's linear optical features are also used in photovoltaic systems, which improve solar cell performance (Hambrock et al., 2001). They are attractive materials for next-generation solar panels because of their tunable bandgap and absorption spectra.

In conclusion, the linear optical characteristics of CdSe quantum dots indicate their tunable and various optical behaviors. Their tailored optical properties, high photoluminescence quantum yields (Ganeev, 2022), and size-dependent emission provide the basis for a broad range of applications, from biological imaging to optoelectronics (Prakash et al., 2002). The use of CdSe quantum dots in revolutionizing numerous fields, promoting creativity and driving technological developments will be further propelled by ongoing research efforts focused on improving synthesis procedures and tackling obstacles.

2.3.2 Nonlinear Optical Properties of CdSe Quantum Dots

Due to their remarkable and versatile properties, CdSe quantum dots have captivated the attention of researchers in the realm of nanotechnology. Researchers in the field of nanotechnology have recently become fascinated by the nonlinear optical features of CdSe quantum dots (Park et al., 1993). This section explores the fascinating field of nonlinear optics in CdSe quantum dots, emphasizing its importance and its uses. When a material's reaction to light is not exactly proportional to the intensity of the input light, this is known as a nonlinear optical phenomenon. The size-dependent electrical characteristics and restricted structure of CdSe quantum dots make for fascinating nonlinear optical activity (Rogalski et al., 2009).

The nonlinear optical absorption and scattering in CdSe quantum dots is one of the most prominent nonlinear optical effects (Klimov et al., Lin and Kuo, 2010). Strong nonlinear absorption features of these dots result in higher-order nonlinear processes and two-photon absorption, among other phenomena (Gao et al., 2011). The quantum confinement effect is essential to these behaviors because it affects the energy band structure and electrical transitions within the dots.

Moreover, because of their small size and distinct electronic structure, CdSe quantum dots exhibit an exceptionally quick response time in their nonlinear optical behaviour. This quick reaction has important implications for nonlinear optical signal processing and ultrafast optical device applications (Lu et al., 2011).

Nonlinear optical properties of CdSe quantum dots enable applications in photovoltaics, optical sensors, and bioimaging (Lim et al., 2011). For instance, their tunable nonlinear optical responses allow them to be used in multiphoton microscopy, providing deeper tissue penetration and better imaging capabilities with better resolution. Furthermore, these dots demonstrate potential for utilizing their nonlinear optical behavior to improve light harvesting and energy conversion efficiency in the development of efficient solar devices (Negi et al., 2013).

However, there are still difficulties in fully comprehending and making use of CdSe quantum dot's nonlinear optical features. For its widespread practical application, issues pertaining to stability, toxicity, and scalability require additional investigation and resolution.

In summary, the nonlinear optical characteristics of CdSe quantum dots offer a fascinating field of study with enormous potential for the progress of new technologies. Their distinct size-dependent behaviour leads to a multitude of applications in photonics and biology, and other domains. Further, research and development are necessary to overcome existing challenges and fully influence the incredible nonlinear optical characteristics of CdSe quantum dots (Goncalves et al., 2023a).

Our goal is focused on understanding and harnessing the nonlinear optical properties of CdSe quantum dots, for their promising future and for further exploration of different paths in this enchanting field.

3. DESCRIPTION OF BROAD AREA

In recent past, an excessive deal of consideration has been paid to studying the size-dependent optical properties of semiconductor nanostructures, like quantum dots (QDs). Quantum size effects cause the properties of semiconductor materials at the nanoscale scale to change. Studying the optical and electrical properties of semiconductor QDs, which are sometimes compared to artificial atoms, is important from a practical standpoint for the creation of different kinds of devices, such as infrared photodetectors (Klimov et al., 2001), lasers, non-volatile memories (Allan and Delerue, 2005), solar cells (Lin et al., 2005), and light-emitting diodes (Peng et al., 2006).

Comparing semiconductor QDs to bulk semiconductors reveals differences in their optical characteristics. The ability to precisely control QD sizes has been made possible by recent advancements in manufacturing techniques, and the use of QDs in devices has revolutionised the semiconductor industry (Hemdana et al., 2013). Semiconductor QDs are attractive because discrete energy levels are possible due to the carrier's confinement in all three dimensions to a nanometer-sized region.

Technological developments in optical spectroscopy have revealed atomically comparable spectra in QDs (Peng et al., 2007). The wavelengths of spectral lines are dependent on dot size because of electronic transitions inside the size-tunable discrete energy levels of QDs. This offers the benefit of continuous spectral tunability over a broad energy range (Irimpan et al., 2008). A flexible method for creating devices based on quantum effects is to engineer semiconductor QD structures. These structures can have distinct electron transport and both linear and non-linear optical properties over a broad spectral range.

Depending on how strong the applied electric field is, a material's optical response to incident radiation might behave either linearly or nonlinearly. A linear relationship between the applied electric field strength and the induced polarization arises when the applied electric field strength is of the order of the internal atomic field strength, leading to linear phenomena. When the applied electric field strength is greater than the internal atomic field strength, nonlinear optical effects develop (Fedd et al., 2017).

In nonlinear optical effects, there is a nonlinear dependence of the induced polarisation on the applied optical field strength. The discovery of numerous nonlinear optical phenomena and the subsequent dramatic shift in optical technology did not occur until after the invention of the laser (Luque et al., 2013). Electronic transitions arising from incident radiation's interaction with matter are the source of optical characteristics. Electronic transitions can appear as interband or intersublevel transitions in quantum dots (QDs).

Interband transitions transpire between the limited states of the valence and conduction bands, whereas intersublevel transitions happen inside the constrained states of the valence or conduction band. Interband transitions in semiconductor nanostructures were the main focus of early research (Peng et al., 2006) (Peng et al., 2007). Transitions within the same band, however, have drawn more attention recently (Germeau et al., 2003) (Boucaud and Sauvage, 2003). QDs with intersublevel transitions have the benefit of having transition energy in the infrared spectrum. Moreover, because only one kind of charge carrier is involved, intersublevel transitions allow for the independent study of hole and electron dynamics.

Numerous linear and nonlinear optical processes, including photoabsorption, photoelectric processes, second harmonic generation, optical rectification, third harmonic generation, intensity-dependent refractive index, sum frequency generation, two-photon bound-bound (TPBB), and two-photon bound-free (TPBF) processes, are based on these transitions (Rabanian et al., 2023c). Through a variety of optical parameters, such as linear and nonlinear optical susceptibilities, linear and nonlinear photoabsorption coefficients, and changes in refractive index, we investigate these optical processes in depth in our work. Until now, the electric dipole approximation has been the basis for most theoretical studies aimed at examining optical characteristics in nanostructures. When the input photon frequency meets the requirements of the dipole approximation and the incident photon wave vector ($\vec{k}$) is in agreement with the position vector ($\vec{r}$) of the electron in the dot, then this approximation can be used. Nevertheless, the dipole approximation is no longer adequate when the radiation wavelength decreases.

For high-frequency photons, the spatial variation of the radiation field must be considered. It is nearly impossible to ignore the higher terms' contribution to multipolar growth for nonlinear cases. The response of the electric quadrupole term to the applied electric field generates electric quadrupole effects (Saar, 1993). When a high-intensity incident electric field interacts with a quantum dot (QD), an electric quadrupole effect is observed (Wagner et al., 2005). Including the electric quadrupole effect modifies absorption spectra, improves optical characteristics, and affects how they depend on the frequency and intensity of the incident radiation.

The production of quantum dots (QDs) involves the use of a wide variety of semiconductor materials, CdSe is emerging as a material of great potential for both linear and nonlinear optical applications (Zurita-Sa Nchez and Novotny, 2002). Comparing semiconductor QDs to bulk semiconductors, their nonlinear optical characteristics are significantly improved (Brunhes et al., 1999). Studying the optical characteristics of semiconductor CdSe QDs is the main goal of this work. This work is inspired by the vital purposes, semiconductor QDs accomplish in numerous fields, including optoelectronics, multiphoton bio-imaging (Caruge et al., 2008), QD based detector and laser creation (Abbas et al., 2019). Due to its physical characteristics that make it ideal for high-power applications, UV, IR, and FTIR emitters, as well as high-temperature electronic devices. Therefore, CdSe QDs are encouraging materials for optical devices and confirm noticeable applications in nonlinear optics (NLO) and other areas as well.

The semiconductor CdSe is especially well-suited for high-temperature, high-power, and short-wavelength optoelectronic applications due to its broad band gap. Due to its direct broad band gap of 1.74 eV and high exciton binding energy of 15-60 meV, CdSe is a suitable material for a wide range of optoelectronic applications (Majdabadi et al., 2015). The absence of inversion symmetry in this material accounts for its even-order nonlinear optical capabilities and also contributes to its piezoelectric features by causing significant spontaneous polarisation.

4. OBJECTIVES OF THE STUDY

The aim of this work is to explore the nonlinear optical properties of CdSe single-electron-charged CdSe (QDs). These properties are arising from the interactions of electric dipole and electric quadrupole interactions induced intersublevel transitions within the conduction band of CdSe QDs. Employing the principles of quantum mechanics and density matrix formulation, we examine these transitions, which are pivotal for comprehending both linear and nonlinear phenomena. Our investigation primarily focuses on nonlinear optical properties of CdSe quantum dots. The work consists of two parts:

4.1 Nonlinear optical properties of CdSe QDs:

The investigation of nonlinear optical properties of CdSe QD will involve analyzing single photon processes and multiphoton processes in terms of:

- Second order nonlinear susceptibility giving rise to second harmonic generation (SHG) and the sum frequency generation (SFG)
- Second order nonlinear susceptibility giving rise to optical rectification (OR).
- Third order nonlinear susceptibility giving rise to the third harmonic generation (THG) and SFG.

4.2 In addition to investigation of nonlinear optical properties, following effects will also be investigated:

- Effect of dot size on the nonlinear optical properties
- Effect of surrounding matrix on the nonlinear optical properties of CdSe QDs
- Effects of nonparabolicity of bands on the nonlinear optical properties of CdSe QDs
- Effect of shape on energy of CdSe QDs.

5. METHODOLOGY TO BE OPTED

The methodology here adopted is the density matrix approach

5.1 Density matrix approach

The optical attributes of a material are elaborately linked to the electronic and molecular composition of the material. Mutually linear and nonlinear susceptibilities serve as metrics for these characteristics. Precise microscopic calculations are crucial for the understanding of these optical susceptibilities. When dealing with scenarios that involve atomic resonant frequencies and the need for reliable results, the density matrix approach turns up as the most appropriate method for calculating optical susceptibilities.

Taking into consideration the system's wave function, we consider $\Psi_s(\tilde{r}, t)$ as the wave function of system under the influence of some applied optical field.

For the system the density matrix element can be expressed as $\widehat{\rho_{nm}} = \overline{C_m^* C_n}$ where the ensemble average is resembled by the overbar.

The wavefunction $\Psi_s(\tilde{r}, t)$ satisfies the Schrodinger equation,

$$i\hbar\frac{\partial\Psi_s(\tilde{r},t)}{\partial t} = H\,\Psi_s(\tilde{r}, t) \tag{6}$$

where $\widehat{H} = \widehat{H_0} + \hat{V}(t)$ is a Hamiltonian operator. The unperturbed Hamiltonian is represented by $\widehat{H_0}$ and the radiation's interaction energy with the system is indicated by $\hat{V}(t)$.

$\widehat{H_0}$ consists the time independent eigen states $u_n(\vec{r})$

Also, $\widehat{H_0}\,u_n(\vec{r}) = E\,u_n(\vec{r})$ (7)

Here $u_n(\vec{r})$ is forming a complete set of orthonormal functions. The wave function $\Psi_s(\tilde{r}, t)$ in equation (6) can also be expanded in terms of $u_n(\vec{r})$ as,

$\Psi_s(\tilde{r}, t) = \sum_n C_n^s(t)u_n(\vec{r})$ (8)

The coefficient in the expansion determines the probability of the system being in energy eigen state n at time t.

The density matrix equation of motion (Liouville equation),

$$\frac{\partial\rho_{nm}}{\partial t} = \frac{1}{\hbar}[\hat{H}, \hat{\rho}]_{nm} - \gamma_{nm}(\rho_{nm} - \rho_{nm}^{(eq)}) \tag{9}$$

$\rho_{nm}^{(eq)} = 0$ for $n \neq m$, assuming $\gamma_{nm} = \gamma_{mn}$

In equation (9) using commutator $[\widehat{H_0}, \hat{\rho}]_{nm} = (\widehat{H_0}\hat{\rho} - \hat{\rho}\,\widehat{H_0})_{nm} = (E_n - E_m)\,\rho_{nm}$

It is obtained as:

$$\frac{\partial\rho_{nm}}{\partial t} = -i\omega_{nm}\rho_{nm} - \frac{i}{\hbar}\sum_v(V_{nv}\rho_{vm} - \rho_{nm}V_{vm}) - \gamma_{nm}(\rho_{nm} - \rho_{nm}^{(eq)}) \tag{10}$$

With the frequency $\omega_{nm} = \frac{E_n - E_m}{\hbar}$ where the energy states of n and m are E_n and E_m respectively.

The analytical solution of the equation (10) is not possible so a perturbative approach is used here. For the perturbative method V_{ij} is expanded in terms of λV_{ij}, λ is the expansion parameter here that necessarily characterizes the perturbation strength which ranges from 0 to 1.

The power series solution of the equation (10) in terms of the λ is,

$$\rho_{nm} = \rho_{nm}^{(0)} + \lambda\rho_{nm}^{(1)} + \lambda^2\rho_{nm}^{(2)} + \ldots\ldots \quad (11)$$

By using the equation (11) and equating the coefficient of each power of λ in equation (10) we obtained,

$$\frac{\partial\rho_{nm}^{(0)}}{\partial t} = -i\omega_{nm}\rho_{nm}^{(0)} - \gamma_{nm}(\rho_{nm}^{(0)} - \rho_{nm}^{(eq)}) \quad (12)$$

$$\frac{\partial\rho_{nm}^{(1)}}{\partial t} = -(i\omega_{nm} + \gamma_{nm})\rho_{nm}^{(1)} + \frac{1}{i\hbar}\left[\hat{V},\ \hat{\rho}^{(0)}\right]_{nm} \quad (13)$$

$$\frac{\partial\rho_{nm}^{(2)}}{\partial t} = -(i\omega_{nm} + \gamma_{nm})\rho_{nm}^{(2)} + \frac{1}{i\hbar}\left[\hat{V},\ \hat{\rho}^{(1)}\right]_{nm} \quad (14)$$

In case of the absence of external field equations the evolution of time for the system. For steady state the solution is $\rho_{nm}^{(0)} = \rho_{nm}^{(eq)}$. Once $\rho_{nm}^{(0)}$ is known the order terms $\rho_{nm}^{(N)}$ can also be obtained by integrating the equations (13 and 14).

$$\rho_{nm}^{(1)}(\mathrm{t}) = \int_{-\infty}^{t}\frac{1}{i\hbar}\left[\hat{V}\,(t'), \widehat{\rho^{(0)}}\right]_{nm} e^{(i\omega_{nm}+\gamma_{nm})(t'-t)}\,\mathrm{dt} \quad (15)$$

The applied field is expressed as,

$$\tilde{\xi}\,(\mathrm{t}) = \sum_p \xi(\omega_p)e^{-i\omega_p t} \quad (16)$$

And $\hat{V}\,(t) = -\vec{\mu}.\vec{\xi}\,(\mathrm{t})$

The polarization can be described as,

$P_i(\boldsymbol{\omega}_p) = \sum_j \mathcal{E}_0\chi_{ij}^{(1)}(\boldsymbol{\omega}_p)\,\boldsymbol{\xi}_j(\omega_p)$ in which the linear susceptibility is defined as,

$$\chi_{ij}^{(1)}\,(\boldsymbol{\omega}_p) = \frac{N}{\mathcal{E}_0\hbar}\sum_{nm}(\rho_{mm}^{(0)} - \rho_{nn}^{(0)})\frac{\mu_{mn}^{i}.\mu_{nm}^{j}}{(\omega_{nm} - \omega_p) - i\gamma_{nm}} \quad (17)$$

If all the population is in the ground level 'a' then $\rho_{aa}^{(0)} = 1$ and $\rho_{mm}^{(0)} = 0$ for $m \neq a$

$$\chi_{ij}^{(1)}\,(\boldsymbol{\omega}_p) = \frac{N}{\mathcal{E}_0\hbar}\sum_n\left[\frac{\mu_{an}^{i}.\mu_{na}^{j}}{(\omega_{na} - \omega_p) - i\gamma_{na}} + \frac{\mu_{na}^{i}.\mu_{an}^{j}}{(\omega_{na} - \omega_p) - i\gamma_{na}}\right]$$

When the frequency $\boldsymbol{\omega}_p$ is in the vicinity of the atomic resonance frequency the second term in the equation (17) can be dropped. Whereas, the imaginary and real part of the susceptibility are related to the absorption coefficient and refractive index respectively.

The second order nonlinear susceptibility is given by,

$$\chi_{ijk}^{(2)}\,(\boldsymbol{\omega}_p + \boldsymbol{\omega}_q;\ \boldsymbol{\omega}_p,\ \boldsymbol{\omega}_q) = \frac{N}{2\mathcal{E}_0\hbar^2} \times$$

$$\sum_{mnv}\left(\rho_{mm}^{(0)}-\rho_{vv}^{(0)}\right)\frac{\mu_{mn}^{i}\mu_{nv}^{j}\mu_{vm}^{k}}{[(\omega_{nm}-\omega_{p}-\omega_{q})-i\gamma_{nm}][(\omega_{vm}-\omega_{p})-i\gamma_{vm}]}+$$
$$\frac{\mu_{mn}^{i}\mu_{nv}^{k}\mu_{vm}^{j}}{[(\omega_{nm}-\omega_{p}-\omega_{q})-i\gamma_{nm}][(\omega_{vm}-\omega_{p})-i\gamma_{vm}]}-\left(\rho_{vv}^{(0)}-\rho_{nn}^{(0)}\right)\frac{\mu_{mn}^{i}\mu_{nv}^{j}\mu_{vm}^{k}}{[(\omega_{nm}-\omega_{p}-\omega_{q})-i\gamma_{nm}][(\omega_{vm}-\omega_{p})-i\gamma_{vm}]}+$$
$$\frac{\mu_{mn}^{i}\mu_{nv}^{k}\mu_{vm}^{j}}{[(\omega_{nm}-\omega_{p}-\omega_{q})-i\gamma_{nm}][(\omega_{vm}-\omega_{p})-i\gamma_{vm}]}$$

the above density matrix approach will be used to study the nonlinear absorption, optical rectification, SHG, SFG for the CdSe QDs.

6. EXPECTED OUTCOMES OF THE RESEARCH

It is intended to obtain enhanced value of nonlinear optical susceptibility associated with different nonlinear optical processes in CdSe QDs as compared to bulk CdSe. Also, it is expected that nonlinear optical properties of CdSe QD depends on dot size, surrounding matrix, shape of the dot and polarization states of incident pump beams. Our study on nonlinear optical properties of CdSe QDs have implications for experimental studies that could significantly improve optical devices such as photodetectors, far infrared laser amplifiers, and high-speed electro-optical modulators. Understanding and exploring these properties are crucial for the advancement of next-generation nanophotonic technologies.

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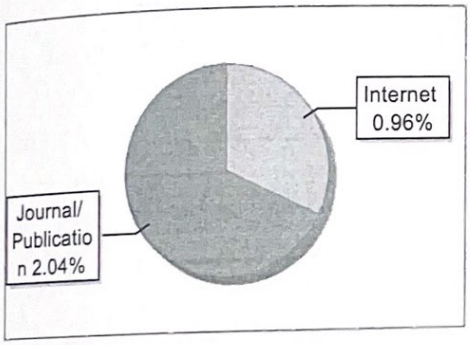

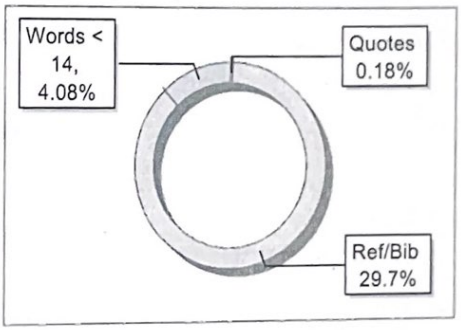

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Electric dipole and quadrupole interaction assisted nonlinear optical properties of CdSe Quantum dot

SHORT SYNOPSIS

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ABSTRACT

Quantum dots (QDs) have received significant attention due to their unique optical properties and potential applications in photonics, optoelectronics, and quantum computing. Our synopsis subject is dedicated to the exploration of some of the nonlinear optical characteristics of CdSe quantum dots, highlighting their distinctive attributes and the mechanisms behind nonlinear optical phenomena. Nonlinear optical effects in quantum dots differ fundamentally from those

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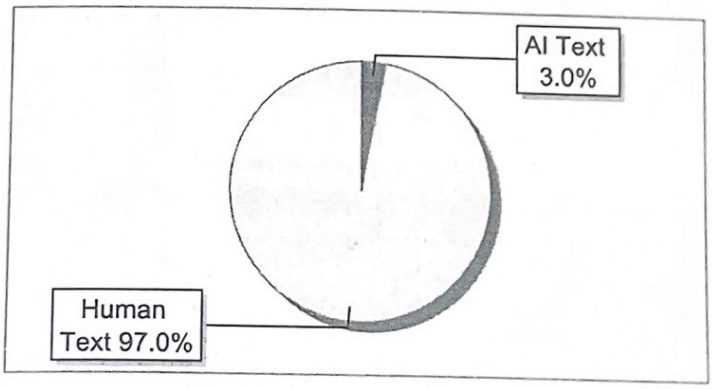

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